Anomalous Exciton-Condensation in Graphene Bilayers

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In ordinary semiconductor bilayers, exciton condensates appear at total Landau level filling factor $\nu_T = 1$. We predict that similar states will occur in Bernal stacked graphene bilayers at many non-zero integer filling factors. For $\nu_T = -3, 1$ we find that the superfluid density of the exciton condensate vanishes and that a finite-temperature fluctuation induced first order isotropic-smectic phase transition occurs when the layer densities are not balanced. These anomalous properties of bilayer graphene exciton condensates are due to the degeneracy of Landau levels with $n = 0$ and $n = 1$ orbital character.

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Introduction— When two semiconductor quantum wells occupied by half-filled Landau levels are narrowly separated, the bilayer system ground state spontaneously establishes inter-layer coherence [1]. These broken symmetry states possess a condensate of pairs (each composed of an electron in one layer and a hole in the other) which opens up an energy gap responsible for a total filling factor $\nu_T = 1$ quantum Hall effect, supports a dissipationless counterflow (excitonic) supercurrent, and is responsible for a wide variety of incompletely understood transport anomalies [2, 3]. Recently an interesting new type of bilayer two-dimensional electron system has become available [4, 5] which consists of two carbon-atom honeycomb-lattice (graphene) layers separated by a fraction of a nanometer. The electronic structure of graphene bilayers is full of surprises [6] because of an interplay between the sublattice pseudospin chirality of each layer [7] and the Bernal stacking arrangement, particularly so in the presence of an external magnetic field. In this structure, one of the two-carbon atom sites in each layer has a near-neighbor in the other layer and one does not. Interlayer hopping drives electrons on the closely spaced sites away from the Fermi level, leaving one low-energy site for carbon π-orbitals in each layer. Because the hopping process between low-energy sites occurs in two steps, via an intermediate high-energy state, it turns out[6] that in the presence of a magnetic field both $n = 0$ and $n = 1$ Landau level orbitals have zero kinetic energy, and that the corresponding wavefunctions are localized in opposite layers for opposite (K and K') graphene valleys [5]. Even though the two-layers are close together and band eigenstates at zero-field are a coherent combinations of individual layer components, the zero-energy strong field states occupy a definite layer.

In the absence of electron-electron and Zeeman interactions, a group of eight degenerate Landau levels (the bilayer octet) develops at the Fermi level of a neutral graphene bilayer because of the degeneracy between $n = 0$ and $n = 1$ states combined with spin and valley degeneracy. The octet is revealed by a jump in the quantized Hall conductivity [4] from $-4(e^2/h)$ to $4(e^2/h)$ when the charge density is tuned across neutrality in moderately disordered samples. When the sample quality is sufficient for interactions to dominate over disorder, quantum Hall effects occur [8, 9, 10] at all intermediate integers. Spontaneous valley-symmetry breaking is expected[9] in all but the $\nu_T = 0$ (half-filled octet) case. Interactions favor states with spontaneous coherence between valleys, and hence between layers, because this broken symmetry does not require charge transfer between layers. In this Letter we illustrate the unique and rich properties of bilayer graphene exciton condensates by concentrating on the simplest case in which the octet is occupied either by single majority-spin ($\nu_T = -3$) or a single minority-spin ($\nu_T = 1$) Landau level, and allowing for an external potential difference Δ_V between the layers. We demonstrate that the superfluid density vanishes at these filling factors, and that a finite-temperature fluctuation induced first order isotropic-smectic phase transition occurs when the layer densities are not balanced. These anomalous properties of bilayer graphene exciton condensates are due to the degeneracy of Landau levels with $n = 0$ and $n = 1$ orbital character. Below we first discuss the mean-field ground state, demonstrating that it exhibits spontaneous inter-layer phase coherence for small Δ_V at the filling factors of interest. We then explain why the superfluid density vanishes and discuss two important consequences, namely that the phonon collective mode has quadratic rather than the expected [11] linear dispersion and that long-wavelength instabilities appear when $\Delta_V \neq 0$. We conclude with some speculations on the experimental implications of our findings.

Octet mean-field theory for unbalanced bilayers— Bilayer graphene's $N = 0$ Landau-level octet is the direct product of three $S = 1/2$ doublets: real spin and *which-layer* pseudospin as in a semiconductor bilayer, and the Landau-level ($n = 0, 1$) pseudospin which is responsible for the new physics we discuss in this paper. The octet degeneracy is lifted by the Zeeman coupling Δ_Z, assumed here to maximize spin-polarization, and by Δ_V which

is defined so that it favors top layer occupation when positive. In graphene bilayers, Δ_V also drives [6, 9] a small splitting of the Landau-level pseudospin that plays a key role in this paper: $\Delta_{LL} = \alpha \Delta_V \hbar\omega_c/\gamma_1$ where $\hbar\omega_c = 2.14\, B[\text{Tesla}]$ meV, $\gamma_1 \sim 400$ meV is the interlayer hopping energy, and $\alpha = +1(-1)$ for top(bottom) layers. The fact that LL 1 has a higher energy than LL 0 in the top layer and a lower energy in the bottom layer will prove to be important.

The octet mean-field Hamiltonian can be separated into single-particle and exchange contributions:

$$\begin{aligned} H_{HF} &= E_{\alpha,n,s}\rho_{\alpha,s,n;\alpha,s,n} \\ &\quad -\frac{1}{g} X^{(\alpha,\alpha)}_{n_1,n_4,n_3,n_2}(0) \left\langle \rho^{\alpha,\alpha}_{s,n_1;r,n_2} \right\rangle \rho^{\alpha,\alpha}_{r,n_3;s,n_4} \\ &\quad -\frac{1}{g} X^{(\alpha,\overline{\alpha})}_{n_1,n_4,n_3,n_2}(0) \left\langle \rho^{\alpha,\overline{\alpha}}_{s,n_1;r,n_2} \right\rangle \rho^{\overline{\alpha},\alpha}_{r,n_3;s,n_4}, \end{aligned} \tag{1}$$

(repeated indices are summed over) where the single particle energy (which includes the Hartree capacitive term) is

$$E_{\alpha,s,n} = -s\frac{\Delta_Z}{2} - \alpha\frac{\Delta_V}{2} + \alpha n \Delta_{LL} + \frac{d}{l_B}\left(\frac{\nu}{2} - \nu_{\overline{\alpha}}\right), \tag{2}$$

and the exchange interactions

$$\begin{aligned} X^{(\alpha,\beta)}_{n_1,n_2,n_3,n_4}(\mathbf{q}) &= \int \frac{d\mathbf{p} l_B^2}{2\pi} \frac{e^{-pd(1-\delta_{\alpha,\beta})}}{p l_B} \\ &\quad \times F_{n_1,n_2}(\mathbf{p}) F_{n_3,n_4}(-\mathbf{p}) e^{i\mathbf{q}\times\mathbf{p} l_B^2}. \end{aligned} \tag{3}$$

In these equations, g is the Landau level degeneracy, $d = 3.337$Å is the interlayer separation, l_B is the magnetic length, $\Delta_Z = g\mu_B B$ is the Zeeman coupling, $n = 0, 1$ are LL indices, $\alpha, \beta = t, b$ for top(bottom) layers, $r, s = 1(-1)$ for up(down) spins and $\overline{t} = b, \overline{b} = t$. All energies are in units of $e^2/\varepsilon l_B$. The average value of the operators $\rho^{\alpha,\beta}_{s,n_1;t,n_2} = \sum_X c^\dagger_{\alpha,s,n_1,X} c_{\beta,t,n_2,X}$ (where $c^\dagger_{\alpha,s,n,X}$ creates an electron with guiding-center X in the Landau gauge and layer α, spin s and Landau level character n) must be determined self consistently by occupying the lowest energy eigenvectors of H_{HF}. The form factors ($F_{00}(\mathbf{q}) = e^{-(ql_B)^2/4}$, $F_{10}(\mathbf{q}) = (iq_x + q_y) l_B e^{-(ql_B)^2/4}/\sqrt{2} = [F_{01}(-\mathbf{q})]^*$ and $F_{11}(\mathbf{q}) = (1 - (ql_B)^2/2) e^{(-ql_B)^2/4}$) reflect the character of the two different quantum cyclotron orbits.

Although an infinitesimal Δ_V would be sufficient to produce complete layer polarization in a non-interacting system, a finite value is required once interactions are accounted for. Spontaneous interlayer phase coherence arises because it is able to lower energy by inducing a gap at the Fermi level even when both layers are partially occupied. When the layer index is viewed as a pseudospin [1], the phase-coherent state corresponds to an $\hat{x} - \hat{y}$ easy-plane ferromagnet and the interlayer phase difference corresponds to the azimuthal magnetization orientation. In this language Δ_V is a hard-direction external field which gradually tilts the magnetization direction toward the $\hat{z}$ direction. We find that for $\Delta_V \geq \Delta_V^*(B)$, with $\Delta_V^*(B)/(e^2/\varepsilon l_B) \approx 0.001$ at $B = 10$T, the system exhibits full charge imbalance with all electrons (all minority spin electrons in the $\nu_T = 1$ case) occupying one layer. The critical Δ_V is given by

$$\Delta_V^* = \frac{e^2}{\varepsilon l_B}\left(\frac{d}{l_B} - \sqrt{\frac{\pi}{2}}\left[1 - e^{d^2/2l_B^2}\mathrm{Erfc}\left[\frac{d}{\sqrt{2}l_B}\right]\right]\right). \tag{4}$$

In the graphene bilayer case, the Δ_V sufficient to achieve full pseudospin polarization is reduced compared to the semiconductor case because the layers are close together. For $\Delta_V \leq \Delta_V^*$, the charge-unbalanced mean-field ground state consists of a full Landau level of states [9, 12, 13] which share partially polarized layer and $n = 0$ Landau-level pseudospinors:

$$|\Psi_{GS}\rangle = \prod_X \left[\cos\left(\frac{\theta_V}{2}\right) c^\dagger_{t,0,X} + e^{i\varphi}\sin\left(\frac{\theta_V}{2}\right) c^\dagger_{b,0,X}\right]|0\rangle, \tag{5}$$

where we have dropped the irrelevant spin degree-of-freedom. In Eq.(5), φ is the spontaneous interlayer phase and $\cos(\theta_V) = \Delta_V/\Delta_V^*(B)$.

Vanishing superfluid density at zero bias— The energy cost of phase gradients is the key property of any superfluid. In normal superfluids, including semiconductor bilayer exciton superfluids, the leading term in a phase-gradient expansion of the energy-density is proportional to $|\nabla\varphi|^2$. We now show that the coefficient which specifies the size of this term, often referred to as the superfluid density, is zero in bilayer graphene exciton condensates at zero bias. We proceed by explicitly constructing a pseudospin wave state in which $\nabla\varphi = q\hat{x}$ is constant:

$$|\Psi_q^{PSW}\rangle = \prod_X \left[\cos\left(\frac{\theta_V}{2}\right) c^\dagger_{t,0,X} + e^{iqX}\sin\left(\frac{\theta_V}{2}\right)\Lambda^\dagger_{q,X}\right]|0\rangle, \tag{6}$$

where $\Lambda^\dagger_{q,X} = u_q c^\dagger_{b,0,X} + v_q c^\dagger_{b,1,X}$ and $u_q^2 + v_q^2 = 1$. In this wavefunction the factor $\exp(iqX)$ is responsible for the phase gradient. The term proportional to v_q in Eq. (6) allows for the crucial possibility, unique to bilayer graphene, of combining the phase gradient with an admixture of the $n = 1$ wavefunction.

In mean-field theory the energy is proportional to the square of the density-matrix. We now show that it is possible to choose v_q so that the density matrix is unchanged to first order in q and that the superfluid density vanishes as a consequence. Since v_q must vanish for $q \to 0$ to reproduce $|\Psi_0\rangle$, we have $v_q \sim q$ and $u_q \sim 1$ to this order. It is then easy to see that the density-matrix component within each layer is unchanged to leading order in q. In quantum Hall bilayer exciton condensates, the superfluid density is due to reduced interlayer exchange energy in the presence of a phase gradient[1, 14], and hence to

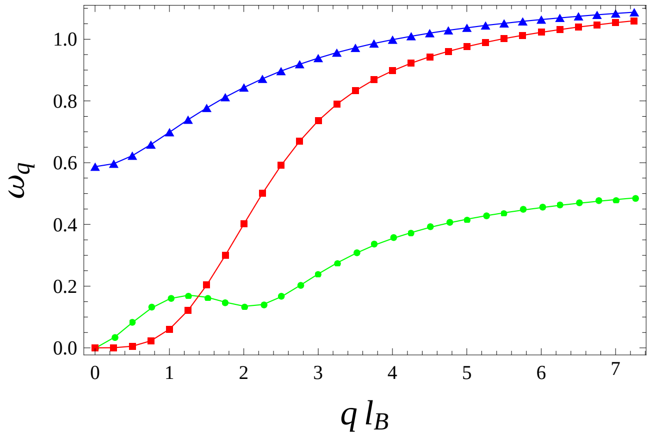

FIG. 1: (Color online) Collective mode dispersion ω_q for balanced Octet QHF in units of interaction strength $e^2/\varepsilon l_B = 11.2(B[T])^{1/2}$ meV as a function of ql_B at a magnetic field of $B = 10$ Tesla. The green (circle) curve shows the inversion symmetric $n = 0$ to $n = 1$ transition mode with $\lim_{q\to 0}\omega_1 \sim q^{3/2}$ dispersion which is unrelated to superfluid properties. The red (box) curve shows the inversion asymmetric superfluid Goldstone mode with the anomalous long-wavelength dispersion $\lim_{q\to 0}\omega_2 \sim q^2$. The blue (triangle) curve is a gapped mode not essential for our discussion (see text for details).

changes in the interlayer density matrix, $\rho_{tb}(\mathbf{r},\mathbf{r}') = \left\langle \Psi_t^\dagger(\mathbf{r})\,\Psi_b(\mathbf{r}')\right\rangle$ where the field operator $\Psi_{t(b)}^\dagger(\mathbf{r}) = 1/\sqrt{L_y}\sum_{n=0,1}\sum_X \phi_n^*(x-X)\, e^{-iXy/l_B^2} c_{t(b),n,X}^\dagger$ with $\phi_n(x)$ the harmonic oscillator state with orbital Landau character n. At zero bias, Eq. (5) with $\varphi = 0$ gives $\rho_{tb}^{GS}(\mathbf{r},\mathbf{r}') = 1/(2L_y)\sum_X \phi_0^*(x-X)\,\phi_0(x'-X)\, e^{-iX(y-y')/\ell_B^2}$ while Eq. (6) implies

$$\rho_{tb}^{PSW}(\mathbf{r},\mathbf{r}') = \frac{1}{2L_y}\sum_X \phi_0^*(x-X)\, e^{-iX(y-y')/l_B^2} e^{iqX} \times\left[u_q\phi_0(x'-X) + v_q\phi_1(x'-X)\right] \quad (7)$$

Since $\phi_1(x) = \sqrt{2}(x/l_B)\phi_0(x)$ and $q(x'-X)$ is small because of the localized oscillator wavefunctions, it follows that $\rho_{tb}^{GS}(\mathbf{r},\mathbf{r}')$ is altered only by an irrelevant phase factor to first order in q if we choose $u_q = 1, v_q = iql_B/\sqrt{2}$. The leading change in $|\rho_{tb}|$ is therefore proportional to q^2 and the leading energy change proportional to q^4.

Unusual collective mode dispersion— We now consider collective excitations of the bilayer quantum Hall exciton condensate at $\nu_T = -3, 1$, first for balanced ($\Delta_V = 0$) bilayers. As discussed above, the ground state is a full Landau level with shared layer symmetric $n = 0$ pseudospin orbitals $|+,0\rangle = (|t,0\rangle + |b,0\rangle)/\sqrt{2}$. The pseudospinor fluctuations that are quantized in the system's collective modes mix in components from the three orthogonal pseudospin states: $|+,1\rangle$ also symmetric in layer indices and $|-,0\rangle$ and $|-,1\rangle$ antisymmetric in layer indices. Fig. 1 shows the collective mode spectrum calculated in the time-dependent mean-field theory [15]. The inversion symmetric mode (transition $|+,0\rangle \to |+,1\rangle$) is unrelated to superfluidity and has been discussed previously [9]. For $q = 0$ the two asymmetric modes correspond respectively to global rotations of the $|+,0\rangle$ pseudospinor toward the states $|-,0\rangle$ and $|-,1\rangle$ respectively. The later rotation costs a finite energy because the exchange energy in $n = 1$ is smaller than in $n = 0$.

One physically transparent way of performing these collective mode calculations is to construct a fluctuation action in which each transition has canonically conjugate density ρ and phase φ components corresponding to the real and imaginary parts of the final state component in the fluctuating spinor. The fluctuation action

$$\mathcal{S}[\rho,\varphi] = \mathcal{S}_B[\rho,\varphi] - \int d\omega \int d^2q\; \mathcal{E}[\rho,\varphi], \quad (8)$$

contains a Berry phase term $\mathcal{S}_B$ [16]

$$\mathcal{S}_B[\rho,\varphi] = \int d\omega \int d^2q \left[\frac{1}{2}\rho_{-\mathbf{q}}^\dagger \mathcal{D}\rho_{\mathbf{q}} - \varphi_{-\mathbf{q}}^\dagger \mathcal{D}^\dagger \varphi_{\mathbf{q}}\right], \quad (9)$$

where $\rho_{\mathbf{q}} = (\rho_{1,\mathbf{q}}, \rho_{2,\mathbf{q}}, \rho_{3,\mathbf{q}})$ and $\mathcal{D} = -i\omega\mathbb{I}_{3\times 3}$, and an energy functional $\mathcal{E}[\rho,\varphi]$ closely related to the discussion in the preceding section

$$\mathcal{E}[\rho,\varphi] = \frac{1}{2}\left[\rho_{-\mathbf{q}}^\dagger\Lambda_\rho(q)\rho_{\mathbf{q}} + \varphi_{-\mathbf{q}}^\dagger\Lambda_\varphi(q)\varphi_{\mathbf{q}}\right]. \quad (10)$$

Here, $\Lambda_\rho(q)$ and $\Lambda_\varphi(q)$ capture the energy cost of small pseudospinor fluctuations and can be evaluated explicitly. Because ρ fluctuations change the charge distribution in the system they remain finite for $q \to 0$ and do not play an essential role in our discussion. At long wavelengths, we find that the inversion asymmetric block in Λ_φ has the form

$$\begin{pmatrix} \frac{X_{1001}^{(+-)}(0)}{2} q^2 l_B^2 + \cdots & i\frac{X_{1001}^{(+-)}(0)}{\sqrt{2}} q l_B + \cdots \\ -i\frac{X_{1001}^{(+-)}(0)}{\sqrt{2}} q l_B + \cdots & X_{1001}^{(+-)}(0) + \cdots \end{pmatrix}, \quad (11)$$

with "$\cdots$" representing terms higher order in ql_B.

In a semiconductor bilayer only $n = 0$ phase fluctuations are possible without paying a kinetic energy penalty. In the strong magnetic field limit the superfluid density is therefore proportional to the interlayer exchange-interaction constant $X_{1001}^{(+-)}$. For the bilayer graphene octet, however, the energy cost of phase variation is proportional to the smallest eigenvalue of the matrix in Eq.(11) and this has the q^4-long wavelength dependence anticipated above. Indeed the eigenvector of this quadratic form captures the orbital character of the Goldstone mode

$$|GM\rangle = |-,0\rangle + i\frac{ql_B}{\sqrt{2}}|-,1\rangle, \quad (12)$$

corresponding to the v_q/u_q ratio in that analysis. We find that the leading small q behavior for the energy functional is

$$\mathcal{E}_{-}[\varphi_{-}] = \beta(ql_B)^4\varphi_{-}^2 + \cdots, \tag{13}$$

where $\beta = 0.093(e^2/\varepsilon l_B)$ at 10T and φ_- is the amplitude of the lowest-energy eigenmode of the phase matrix $\Lambda_\varphi(q)$. Because of the conjugate relationship between ρ and φ fluctuations, the collective mode energy is proportional to the square root of the eigenvalues of Λ_φ and Λ_ρ so that the quadratic Goldstone mode dispersion simply signals the vanishing superfluid density.

Long-wavelength instability at finite bias— The q^4 interaction energy cost of inter-layer phase gradients holds for balanced and unbalanced bilayers. In unbalanced layers, however, the Landau level splitting Δ_{LL} means that the single-particle energy is lowered by transitions to the $n = 1$ Landau level in the bottom layer. For unbalanced layers we find that the phase fluctuation action goes to

$$\begin{pmatrix} \frac{X^{(+-)}_{1001}(0)}{2}q^2l_B^2 + \cdots & i\frac{X^{(+-)}_{1001}(0)}{\sqrt{2}}ql_B + \cdots \\ -i\frac{X^{(+-)}_{1001}(0)}{\sqrt{2}}ql_B + \cdots & X^{(+-)}_{1001}(0) - \cos(\theta_V)\,\Delta_{LL} + \cdots \end{pmatrix}. \tag{14}$$

The energy cost of phase fluctuations is again proportional to the smallest eigenvalue of the phase matrix,

$$\mathcal{E}_{-}[\varphi_{-}] = -\frac{1}{2}\cos(\theta_V)\,\Delta_{LL}(ql_B)^2\varphi_{-}^2 + \beta(ql_B)^4\varphi_{-}^2 + \cdots, \tag{15}$$

which is negative at small q, indicating that a uniform condensate is unstable when $\Delta_{LL} \neq 0$. The energy functional in Eq.(15) reduces to the classical Swift-Hohenberg (SH) model [17] Hamiltonian:

$$\mathcal{E}[\varphi_{-}] = \left[\frac{\Delta_0}{2} + \frac{\xi_0^4}{2}(\nabla^2 + q_0^2)^2\right]\varphi_{-}^2 + \frac{\lambda}{4!}\varphi_{-}^4, \tag{16}$$

when we set $\Delta_0 = -(\cos(\theta_V)\,\Delta_{LL})^2/8\beta$, $\xi_0^2 = \sqrt{2\beta}l_B^2$ and $(q_0l_B)^2 = \cos(\theta_V)\,\Delta_{LL}/4\beta$. Using the detailed microscopic form of the mean-field energy functional, we estimate that $\lambda \sim (q_0^4l_B^2)(e^2/\varepsilon l_B)$. The SH model exhibits a fluctuation-induced first order smectic-isotropic phase transition from a stripe ordered phase with $\langle\varphi_-\rangle = A\cos(\mathbf{q}_0\cdot\mathbf{r})$ to a disordered phase with $\langle\varphi_-\rangle = 0$. Following the self-consistent Hartree approximation analysis[17] of the Swift-Hohenberg model, for $\Delta_V < \Delta_V^*$ we estimate the transition temperature to be

$$k_BT_c = \frac{4\xi_0^2}{2.03\lambda}|\Delta_0|^{3/2} = 1.97\frac{\beta}{e^2/\varepsilon l_B}\left(\frac{\hbar\omega_c}{\gamma_1}\Delta_V^*\right)\left(\frac{\Delta_V}{\Delta_V^*}\right)^2. \tag{17}$$

which is typically below 10mK.

Discussion— We identify the $T > T_c$ phase with a normal quantum Hall ferromagnet and the $T < T_c$ phase with a quantum Hall smectic state which should exhibit anisotropic transport properties. For $T > T_c$ we expect properties similar to those observed in semiconductor bilayers[2] except that the coherent interlayer tunneling processes, which plays a key role in tunneling experiments, should be essentially absent. In graphene bilayers therefore, spontaneous coherence is likely to be most conveniently manifested by strongly enhanced Hall drag. Finally we note that trigonal warping terms, which we have neglected, will reduce Δ_{LL}[9] by at most 5% for magnetic fields of interest, that correlation effects we have not considered could contribute positively or negatively to the superfluid density, and that the small superfluid densities in this system might lead to important thermal fluctuation effects beyond those considered here. This work was supported by the NSF under grant DMR-0606489 (AHM) a grant by NSERC (RC) and the State of Florida (YB).

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